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A FORMAL SOLUTION OF LIOUVILLE'S EQUATION

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ABSTRACT

A formal solution of Liouville's equation for both the classical and the quantum mechanical case is presented. The derivation closely follows the approach employed by Feynman in his papers on the theory of positrons in *The Physical Review*, Volume 76, pages 749, 769, 1949. A scattering operator S is found which connects the distribution function at time t' with the distribution function at any later time t. Using a diagram, each term of this scattering operator can be represented uniquely and conveniently. The topological structure of these diagrams is the same in the classical as well as in the quantum mechanical case.

I. INTRODUCTION

The Liouville equation represents a natural starting point for studies of non-equilibrium statistical mechanics (see Ref. 1-7). The many-body problem encountered in statistical mechanics may advantageously be handled by the mathematical methods especially developed for the infinite many-body problem of quantum field theory (see Ref. 8). As an example, Prigogine and collaborators (see Ref. 9) used the S matrix formalism in some of their papers. Furthermore, the well-known approach of Prigogine and collaborators to the solution of Liouville's equation by means of Fourier transforms and a diagram technique (see Ref. 10) bears much formal resemblance to field theoretical methods.

In this Report a formal solution of the Liouville equation is presented; the solution is based on Feynman's approach to the solution of the Schrodinger equation (see Ref. 11). The actual derivation is given in Section II. An integral kernel plays the central role of the development. This kernel is somewhat similar to the phase-space transformation function introduced by Ross and Kirkwood (see Ref. 12), although it is a Green's function rather than a solution of the

homogeneous Liouville equation. The various contributions of a perturbation expansion of this kernel give rise to corresponding contributions to the distribution function which may be classified by diagrams.

The modifications due to quantum mechanics are also outlined for the case of the Wigner distribution function (see Ref. 13). The physical significance of the diagrams is clarified in Section III by means of an equivalent perturbation expansion of the equations of motion.

II. DERIVATION

The classical Liouville equation for an N-particle system may be written as

$$\left(\frac{\partial}{\partial t} + \mathbf{V} \cdot \nabla_{\mathbf{R}}\right) f(\mathbf{R}, \mathbf{V}, t) = -\mathbf{F}(\mathbf{R}, \mathbf{V}) \cdot \nabla_{\mathbf{V}} f(\mathbf{R}, \mathbf{V}, t) \tag{1}$$

where $\mathbf{R}$ is the abbreviation for the set of position coordinates $\mathbf{r}_1, \mathbf{r}_2, \cdots, \mathbf{r}_N$ of the N particles. Similarly $\mathbf{V}$ stands for $\mathbf{v}_1, \mathbf{v}_2, \cdots \mathbf{v}_N$ the velocities of the particles. $\mathbf{F} \equiv \mathbf{F}_1, \mathbf{F}_2, \cdots, \mathbf{F}_N$, where $\mathbf{F}_j$ is the force acting on particle j divided by its mass. $\mathbf{F}$ may be a function of both position and velocity. The symbol $\mathbf{V} \cdot \nabla_{\mathbf{R}}$ is an abbreviation for

$$\mathbf{V} \cdot \nabla_{\mathbf{R}} = \sum_{i=1}^{N} \mathbf{v}_i \cdot \nabla_{\mathbf{r}_i}$$

A kernel $G(\mathbf{R}, \mathbf{V}, t; \mathbf{R}', \mathbf{V}', t')$ is now introduced which allows for the expression of the distribution function $f(\mathbf{R}, \mathbf{V}, t)$ for $t > t'$ by

$$f(\mathbf{R}, \mathbf{V}, t) = \int d\mathbf{R}'\, d\mathbf{V}'\, G(\mathbf{R}, \mathbf{V}, t; \mathbf{R}', \mathbf{V}', t')\, f(\mathbf{R}', \mathbf{V}', t') \tag{2}$$

if it is known at an earlier time t'. The integration runs over the complete phase space $d\mathbf{R}'\, d\mathbf{V}' = d^3r_1'\ d^3r_2' \cdots d^3v_1'\ d^3v_2' \cdots$. The kernel G has the following properties:

$$\left(\frac{\partial}{\partial t} + \mathbf{V} \cdot \nabla_{\mathbf{R}} + \mathbf{F} \cdot \nabla_{\mathbf{V}}\right) G(\mathbf{R}, \mathbf{V}, t; \mathbf{R}', \mathbf{V}', t') = \delta(\mathbf{R} - \mathbf{R}')\ \delta(\mathbf{V} - \mathbf{V}')\ \delta(t - t') \tag{3}$$

$$\lim_{t \to t'} G(\mathbf{R}, \mathbf{V}, t; \mathbf{R}', \mathbf{V}', t') = \delta(\mathbf{R} - \mathbf{R}')\ \delta(\mathbf{V} - \mathbf{V}') \tag{4}$$

Equation (3) shows that G is a Green's function of the Liouville equation. Equation (4) is necessary for Eq. (2) to be consistent. Equations (2), (3) and (4) are strongly reminiscent of Feynman's approach to the solution of Schrodinger's equation. In fact, if the operator $\mathbf{V} \cdot \mathbf{V}_R + \mathbf{F} \cdot \mathbf{V}_V$ is replaced by the Hamiltonian H and the distribution function f is replaced by the wave function ψ, Eqs. (2), (3) and (4) are identical with Feynman's theory. (For the account of this approach see Ref. 12.) It is now proposed to solve Eq. (3) by an iteration method assuming weak interactions. We obtain

$$G = G_0 + G_1 + G_2 + \cdots \tag{5}$$

$$\left(\frac{\partial}{\partial t} + \mathbf{V} \cdot \nabla_{\mathbf{R}}\right) G_0 = \delta(\mathbf{R} - \mathbf{R}')\ \delta(\mathbf{V} - \mathbf{V}')\ \delta(t - t') \tag{6}$$

$$\left(\frac{\partial}{\partial t} + \mathbf{V} \cdot \nabla_{\mathbf{R}}\right) G_n = -\mathbf{F} \cdot \nabla_{\mathbf{V}} G_{n-1} \qquad (n \geq 1) \tag{7}$$

The solution of Eq. (6) is immediately written[1] as

$$G_0(\mathbf{R}, \mathbf{V}, t; \mathbf{R}', \mathbf{V}', t') = \delta(\mathbf{V} - \mathbf{V}')\ \delta[\mathbf{R} - \mathbf{R}' - \mathbf{V}'(t - t')]\ S(t - t') \tag{8}$$

introducing the step function

$$S(x) = \begin{cases} 1 \text{ for } x > 0 \\ 0 \text{ for } x < 0 \end{cases} \tag{9}$$

[1]The solution Eq. (8) is the retarded Green's function. The advanced Green's function is found by replacing $S(t - t')$ by $-S(t' - t)$.

It is easily seen that Eq. (4) is satisfied for expression (8). From Eq. (7) it is apparent

$$G_n(\mathbf{R},\mathbf{V},t;\mathbf{R}',\mathbf{V}',t') = -\int dt''\,d\mathbf{R}''\,d\mathbf{V}''\,G_0(\mathbf{R},\mathbf{V},t;\mathbf{R}'',\mathbf{V}'',t'') \times \mathbf{F}(\mathbf{R}'',\mathbf{V}'')\cdot\nabla_{\mathbf{V}''}\,G_{n-1}(\mathbf{R}'',\mathbf{V}'',t'';\mathbf{R}',\mathbf{V}',t') \tag{10}$$

Successive application of Eq. (10) reveals after straightforward calculation that

$$\begin{aligned} G_n(\mathbf{R},\mathbf{V},t;\mathbf{R}',\mathbf{V}',t') = {} & \int_{t'}^{t} dt_1 \int_{t'}^{t_1} dt_2 \cdots \int_{t'}^{t_{n-1}} dt_n \int d\mathbf{V}_2\,d\mathbf{V}_3\cdots d\mathbf{V}_n \\ & \times (-1)^n\ \delta\left[\mathbf{R}-\mathbf{V}(t-t_1)-\sum_{j=2}^{n}\mathbf{V}_j(t_{j-1}-t_j)-\mathbf{V}'(t_n-t')\right] \\ & \times \mathbf{F}[\mathbf{R}-\mathbf{V}(t-t_1),\mathbf{V}]\cdot\nabla_{\mathbf{V}}\ \delta(\mathbf{V}-\mathbf{V}_2) \\ & \times \mathbf{F}[\mathbf{R}-\mathbf{V}(t-t_1)-\mathbf{V}_2(t_1-t_2),\mathbf{V}_2]\cdot\nabla_{\mathbf{V}_2}\ \delta(\mathbf{V}_2-\mathbf{V}_3)\cdots \\ & \times \mathbf{F}\left[\mathbf{R}-\mathbf{V}(t-t_1)-\sum_{j=2}^{n}\mathbf{V}_j(t_{j-1}-t_j),\mathbf{V}_n\right]\cdot\nabla_{\mathbf{V}_n}\ \delta(\mathbf{V}_n-\mathbf{V}') \end{aligned} \tag{11}$$

Inserting the expressions (8) and (11) into Eq. (2) and performing the phase-space integrations as indicated has the following result. The distribution function at time t is connected with the initial distribution function at time t' through a scattering operator S

$$f(\mathbf{R},\mathbf{V},t) = S\,f[\mathbf{R}-\mathbf{V}(t-t'),\mathbf{V},t'] \tag{12}$$

where

$$S = \sum_{n=0}^{\infty} S_n \qquad (S_0 = 1) \tag{13}$$

and for $n \geq 1$

$$S_n = (-1)^n \int_{t'}^{t} dt_1 \int_{t'}^{t_1} dt_2 \cdots \int_{t'}^{t_{n-1}} dt_n \prod_{\alpha=1}^{n} F[\mathbf{R}-\mathbf{V}(t-t_\alpha);\mathbf{V}]\cdot\mathbf{P}_\alpha \tag{14}$$

with

$$P_\alpha = \nabla_V - (t_\alpha - t')\nabla_R - \sum_{\beta=\alpha+1}^{n} [\nabla_V - (t_\alpha - t_\beta)\nabla_R] = P_{\alpha f} + \sum_{\beta=\alpha+1}^{n} P_{\alpha\beta} \tag{15}$$

The gradient ∇_V in Eq. (15) is always connected with a gradient ∇_R in the specific configuration

$$P_{\alpha\beta} = \nabla_V - (t_\alpha - t_\beta)\nabla_R \tag{16}$$

The operator $P_{\alpha\beta}$ is defined as acting only on that function of R and V which contains the same time label β. Furthermore, ∇_V does not act on the argument $R - V(t - t_\beta)$. In other words

$$P_{\alpha\beta} F[R - V(t - t_\gamma), V] = \delta_{\beta\gamma} \left\{ \overrightarrow{\nabla_V F[R - V(t - t_\gamma), V]} - (t_\alpha - t_\beta) \overrightarrow{\nabla_R F[R - V(t - t_\gamma), V]} \right\} \tag{17}$$

indicating by an arrow on which argument the gradient operates. Inserting Eq. (15) into (14) it is seen that S_n can be expressed by a sum of terms of the following structure

$$S_n = (-1)^n \sum_\gamma \int_{t'}^{t} dt_1 \int_{t'}^{t_1} dt_2 \cdots \int_{t'}^{t_{n-1}} dt_n \sum_{\alpha=1}^{n} F[R - V(t - t_\alpha), V] \cdot P_{\alpha\gamma_\alpha} \tag{18}$$

In Eq. (18) the condition is $\gamma_\alpha > \alpha$ but otherwise arbitrary and the sum runs over all $n!$ possibilities to pick a set of $\gamma_\alpha > \alpha$. Any particular term of the sum may be represented by a diagram. We define the diagrams in the following way: An nth order diagram consists of $n + 1$ vertices labeled with a time coordinate t_α starting with the latest time t_1 on the left (Fig. 1a) and ending with a vertex associated with the earliest time, t' the initial time, on the right. Starting from each vertex α (except the one associated with t') there is one and only one directed solid line which ends at any arbitrary vertex β with an earlier time including the last vertex (Fig. 1b). The last vertex associated with the initial time t' is called the external vertex (each diagram has only one external vertex) all the others are called internal vertices. A solid line starting from any internal vertex and ending at any other internal vertex is called an internal line. If it ends on the external vertex however it is called an external line. Figure 1c shows a possible fifth order diagram with two external and three internal lines; similarly, Fig. 1d shows a diagram with three external and two internal lines. The number of lines is equal to the number of internal vertices;

indicating therefore the order of the diagram. There are $n!$ different diagrams of order n corresponding to the $n!$ different ways of connecting any two of the $n + 1$ vertices by a solid line such that only one solid line starts from any internal vertex. We associate with each vertex α the factor $-\boldsymbol{F}[\boldsymbol{R} - \boldsymbol{V}(t - t_\alpha), \boldsymbol{V}]$ and with an internal line the operator $\boldsymbol{P}_{\alpha\beta}$ which acts in the manner specified by Eq. (17) on the $-\boldsymbol{F}[\boldsymbol{R} - \boldsymbol{V}(t - t_\beta), \boldsymbol{V}]$ associated with vertex β. Furthermore, associated with an external line starting at the vertex α is the operator

$$\boldsymbol{P}_{\alpha/} = \nabla_{\boldsymbol{V}} - (t_\alpha - t')\nabla_{\boldsymbol{R}} \tag{16a}$$

which acts on the initial distribution function represented by the external vertex. We now see that any given diagram (Fig. 1c, d for example) represents uniquely one term of the sum (18). As an example, Fig. 2 shows the three lowest-order diagrams.

According to the rules outlined above the first-order contribution (Fig. 2a) is given by

$$\begin{aligned} S_1 &= -\int_{t'}^{t} dt_1\, \boldsymbol{F}[\boldsymbol{R} - \boldsymbol{V}(t - t_1), \boldsymbol{V}] \cdot \boldsymbol{P}_{1/} \\ &= -\int_{t'}^{t} dt_1\, \boldsymbol{F}[\boldsymbol{R} - \boldsymbol{V}(t - t_1), \boldsymbol{V}] \cdot [\nabla_{\boldsymbol{V}} - (t_1 - t')\nabla_{\boldsymbol{R}}] \end{aligned} \tag{19}$$

The second-order contribution (Fig. 2b) is given by

$$S_2 = \int_{t'}^{t} dt_1 \int_{t'}^{t_1} dt_2\, \boldsymbol{F}[\boldsymbol{R} - \boldsymbol{V}(t - t_1), \boldsymbol{V}] \cdot \boldsymbol{P}_{12}\, \boldsymbol{F}[\boldsymbol{R} - \boldsymbol{V}(t - t_2), \boldsymbol{V}] \cdot \boldsymbol{P}_{2/} \tag{20}$$

Here $\boldsymbol{P}_{12}$ acts on the succeeding $\boldsymbol{F}$ whereas $\boldsymbol{P}_{2/}$ acts on the initial distribution function as explained above. The other second-order contribution (Fig. 2c) is given by

$$S_2' = \int_{t'}^{t} dt_1 \int_{t'}^{t_1} dt_2\, \boldsymbol{F}[\boldsymbol{R} - \boldsymbol{V}(t - t_1), \boldsymbol{V}] \cdot \boldsymbol{P}_{1/}\, \boldsymbol{F}[\boldsymbol{R} - \boldsymbol{V}(t - t_2), \boldsymbol{V}] \cdot \boldsymbol{P}_{2/} \tag{21}$$

where both $\boldsymbol{P}$ act on the initial distribution function.

Turning now to the quantum-mechanical Liouville equation for the Wigner distribution function (see Ref. 15), discussion is restricted to the case in which the force $\boldsymbol{F}$ is derived from

a potential $\boldsymbol{F}(R) = -\nabla_{\boldsymbol{R}}\phi(R)$. The Liouville equation for the Wigner distribution function $f^{(W)}$ is well known (see Ref. 16) to be

$$\left(\frac{\partial}{\partial t} + \boldsymbol{V}\cdot\nabla_{\boldsymbol{R}}\right) f^{(W)} = \frac{2}{\hbar}\sin\left(\frac{\hbar}{2M}\nabla_{\boldsymbol{R}}\cdot\nabla_{\boldsymbol{V}}\right)\phi(\boldsymbol{R})\, f^{(W)} \tag{22}$$

Here the operator $\nabla_{\boldsymbol{R}}$ acts only on the potential $\phi(\boldsymbol{R})$. The sine operator is defined by its power series expansion. It is easily seen that the scattering operator S for $f^{(W)}$ may still be determined by the same diagrams as were used in the classical case. Only the rules of associating an internal vertex and an internal line to a given mathematical quantity have to be changed. An internal vertex α now represents

$$\frac{2}{\hbar}\sin\left(\frac{\hbar}{2M}\nabla_{\boldsymbol{R}}\cdots\right)\phi[\boldsymbol{R} - \boldsymbol{V}(t - t_\alpha)] \tag{23}$$

An internal solid line connecting the vertices α and β represents

$$\boldsymbol{P}'_{\alpha\beta} = -(t_\alpha - t_\beta)\nabla_{\boldsymbol{R}} \tag{24}$$

where $\nabla_{\boldsymbol{R}}$ acts on the potential associated with vertex β. The operator $\boldsymbol{P}_{\alpha\beta}$ of Eq. (16) is also replaced by $\boldsymbol{P}'_{\alpha\beta}$ Eq. (24) in the classical case if the forces do not depend explicitly on the velocity as is seen from Eq. (17). According to Eq. (23) and (24), the complete expression for the part of the diagram shown in Fig. 1b is

$$\cdots\ \frac{2}{\hbar}\sin\left(\frac{\hbar}{2M}\nabla_{\boldsymbol{R}}\cdot\boldsymbol{P}'_{24}\right)\phi[\boldsymbol{R} - \boldsymbol{V}(t - t_2)]\ \cdots \tag{25}$$

An external solid line is still represented by Eq. (16a). As an example, Fig. 2b may be considered, which represents

$$S'_2 = \int_{t'}^{t} dt_1 \int_{t'}^{t_1} dt_2\, \frac{2}{\hbar}\sin\left(\frac{\hbar}{2M}\nabla_{\boldsymbol{R}}\cdot\boldsymbol{P}'_{12}\right)\phi[\boldsymbol{R} - \boldsymbol{V}(t - t_1)]\, \frac{2}{\hbar}\sin\left(\frac{\hbar}{2M}\nabla_{\boldsymbol{R}}\cdot\boldsymbol{P}_{2/}\right)$$

$$\times\ \phi[\boldsymbol{R} - \boldsymbol{V}(t - t_2)]$$

III. DISCUSSION

The rules which generate the scattering operator S being established, the consequences at this point are interesting. First of all, it may be seen that the nth order term of the series for S consists of a sum over n diagrams with a varying number of internal and external lines. The physical significance of an internal or external line will be clarified by the following considerations. In the zeroth order the motion of the particles is undisturbed. They proceed along straight lines. In fact the scattering operator being $S_0 = 1$ to the zeroth order, Eq. (12) becomes

$$f(\boldsymbol{R}, \boldsymbol{V}, t) = f[\boldsymbol{R} - \boldsymbol{V}(t - t'), \boldsymbol{V}, t'] \tag{26}$$

To see how precisely the higher-order terms of the scattering operator introduce deviations from the unperturbed straight lines of the zeroth order approximation, a perturbation scheme applied directly to the equations of motion is outlined which is completely equivalent to the perturbation analysis for the scattering operator described in Sec. II. Confining ourselves to the classical equations of motion and to forces which depend only on the position, we have

$$\ddot{\boldsymbol{R}} = \lambda \boldsymbol{F}(\boldsymbol{R}) \tag{27}$$

together with the initial conditions

$$\boldsymbol{R}(t') = \boldsymbol{R}' \qquad \dot{\boldsymbol{R}}(t') = \boldsymbol{V} \tag{27a}$$

A small expansion parameter λ is introduced

$$\boldsymbol{R} = \boldsymbol{R}_0 + \lambda \boldsymbol{R}_1 + \lambda^2 \boldsymbol{R}_2 + \cdots \tag{28}$$

Inserting Eq. (28) into Eq. (27) gives the successive approximations

$$\begin{aligned} \ddot{\boldsymbol{R}}_0 &= 0 \\ \ddot{\boldsymbol{R}}_1 &= \boldsymbol{F}(\boldsymbol{R}_0) \\ \ddot{\boldsymbol{R}}_2 &= \boldsymbol{R}_1 \cdot \nabla_{\boldsymbol{R}_0} \boldsymbol{F}(\boldsymbol{R}_0) \end{aligned} \tag{29}$$

and the initial conditions are

$$R_0(t') = R' \qquad \dot{R}_0(t') = V$$

$$R_1(t') = \dot{R}_1(t') = 0 \tag{29a}$$

$$R_2(t') = \dot{R}_2(t') = 0$$

It follows from Eq. (29) and (29a) that the successive approximations to the acceleration are given by

$$\ddot{R}_0 = 0 \tag{30a}$$

$$\ddot{R}_1 = F[R' - V(t' - t)] \tag{30b}$$

$$\ddot{R}_2 = \int_{t'}^{t} dt_1 \int_{t'}^{t_1} dt_2 \, F[R' - V(t' - t_2)] \cdot \nabla_R F[R' - V(t' - t)] \tag{30c}$$

A glance back to Eq. (19), (20), and (21) and a comparison with Eq. (30b) and (30c) reveals that Eq. (19), the first-order scattering operator, takes into account a deviation from the unperturbed straight paths of the particles to exactly the same degree of approximation on which Eq. (30b) is based. Equation (20), corresponding to the diagram with one internal line (Fig. 2b), takes into account a first-order correction to the first-order effect given by Eq. (19), and it is therefore of second order. The exact analog to this correction, expressed by an internal line, is Eq. (30c). But this is not all that might happen in second order! Actually, a new "scattering" may be introduced at some other time between t' and t. But Eq. (21), corresponding to the diagram with two external lines (Fig. 2c), represents precisely the contribution from this event again to the correct order of magnitude.

These findings may now immediately be generalized to an nth order contribution. Suppose there is a specific nth-order diagram which represents one of the $n!$ nth order contributions to the scattering operator. This diagram generates $n + 1$ diagrams of the $(n + 1)$st order. By adding a new internal vertex to the left of the nth-order diagram and connecting it to either of the n other internal vertices n new possible diagrams with one more internal line is obtained. Connecting it with the external vertex, one new diagram with one more external line is obtained. From the previous discussion it is clear that the $(n + 1)$st order diagrams generated from a specific nth-order diagram take into account a correction to the path of any one particle in the sense of Eq. (30c). This is possible in n different ways corresponding to the n different new internal lines, whereas the possibility of a new scattering is represented by the addition of a new external line. In short, internal lines represent corrections to already existing scatterings and external lines represent the introduction of new scatterings. Of course, it should be realized

that this explanation is more or less heuristic, since there is no explicit introduction of scattering cross-sections. The scattering operator, rather, gives the detailed time dependence of the distribution function, and is therefore completely equivalent to the exact knowledge of the orbits of all the particles involved. This is far too much information to be useful. In fact, to extract useful information averaging procedures must be employed. Singlet, doublet, etc. distribution functions may be introduced, and subsequently, Kirkwood's coarse-graining device (see Ref. 17), "Stosszahlansatz," etc. may be used. However, this is beyond the scope of the present Report.

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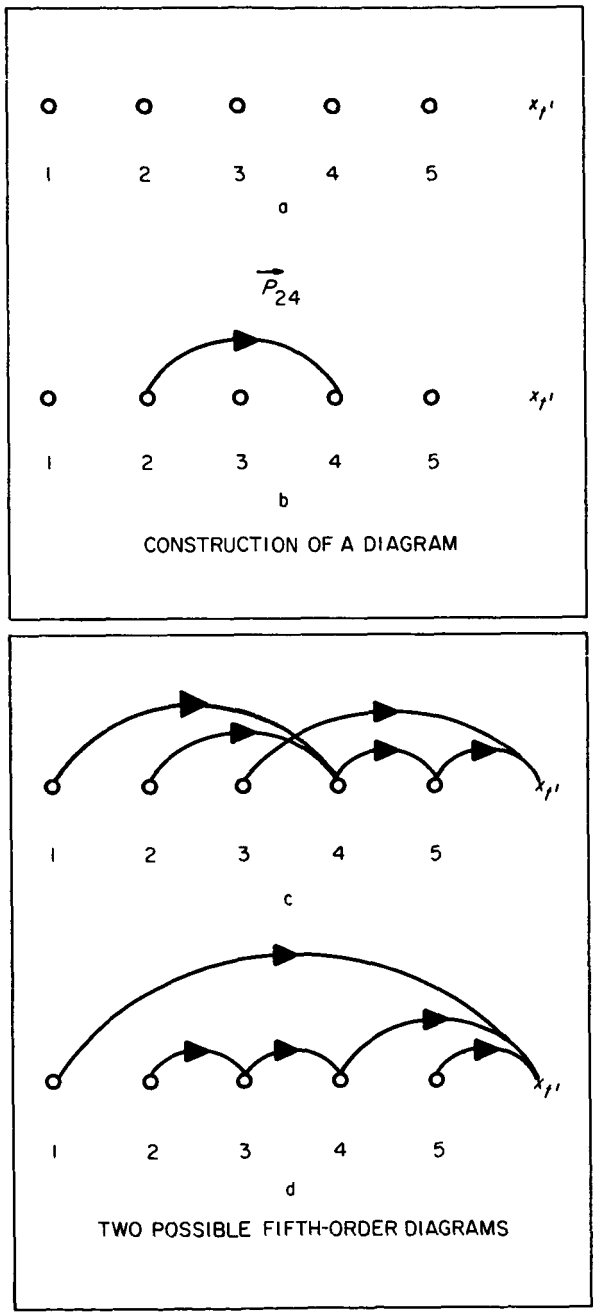

Fig. 1. Construction and Examples of Fifth-Order Diagrams

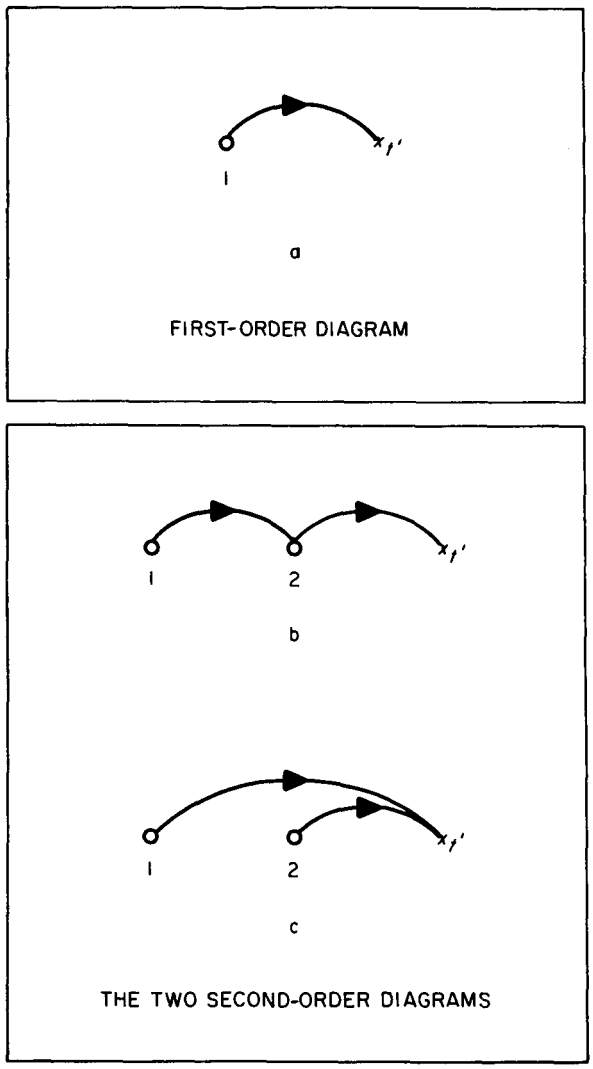

Fig. 2. First and Second-Order Diagrams